

SOLID HIGH POLYMER ELECTROLYTE FUEL CELL

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Abstract of JP2000188111

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(54) SOLID HIGH POLYMER ELECTROLYTE FUEL CELL

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CLAIMS

[Claim(s)]

[Claim 1] Said ion exchange resin contained in said air pole in the solid-state polyelectrolyte mold fuel cell with which the gas diffusion electrode containing a catalyst and ion exchange resin was made into the fuel electrode and the air pole, said fuel electrode had been arranged at one side of a film-like solid-state polyelectrolyte, and said air pole has been arranged in another field, respectively is a solid-state polyelectrolyte mold fuel cell characterized by being ion exchange resin which consists of a copolymer including the following polymerization unit A, the following polymerization unit B, and the following polymerization unit C.

Polymerization unit A: The polymerization unit based on the perfluoro vinyl ether which does not have the polymerization unit based on tetrafluoroethylene, the polymerization unit based on the perfluoro vinyl ether which has a polymerization unit B:sulfonic group, a polymerization unit C:ion exchange group, or the radical of the precursor.

[Claim 2] said copolymer — said polymerization unit B and said polymerization unit C — a total amount — said polymerization unit A — receiving — 10-25-mol% — the solid-state polyelectrolyte mold fuel cell according to claim 1 contained.

[Claim 3] said copolymer — said polymerization unit B — the total amount of said polymerization unit A and said polymerization unit C — receiving — more than 5mol% — the solid-state polyelectrolyte mold fuel cell according to claim 2 contained.

[Claim 4] said copolymer — said polymerization unit C — the total amount of said polymerization unit A and said polymerization unit B — receiving — more than 2mol% — the solid-state polyelectrolyte mold fuel cell according to claim 3 contained.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001] [Field of the Invention] This invention relates to a solid-state polyelectrolyte mold fuel cell.

[0002] [Description of the Prior Art] Hydrogen and an oxygen fuel cell attract attention as a generation-of-electrical-energy system which the resultant of is only water theoretically and does not almost have an adverse effect to earth environment. Although the solid-state polyelectrolyte mold fuel cell was once carried in the spacecraft in the Project Gemini and the biotechnology satellite plan, cell output density of those days was low. Then, the alkaline fuel cells of high performance are developed more, and alkaline fuel cells are adopted as space until it results in a current space shuttle.

[0003] However, the solid-state polyelectrolyte mold fuel cell attracts attention again by advance of a technique in recent years. The following two points are mentioned as the reason.

(1) The film of high conductivity was developed as a solid-state polyelectrolyte. (2) Very big activity came to be acquired by supporting to carbon the catalyst used for a gas diffusion electrode layer, and covering this with ion exchange resin further.

[0004] And many examination is made about the manufacture approach of of the electrode and solid-state polyelectrolyte film zygote of a solid-state polyelectrolyte mold fuel cell (only henceforth a zygote). Since the solid-state polyelectrolyte mold fuel cell by which current examination is carried out has operating temperature as low as 50-120 degrees C, it has the fault which exhaust heat cannot use effectively for the auxiliary machine power of a fuel cell etc. easily. Output density with an especially high solid-state polyelectrolyte mold fuel cell is demanded also in the semantics with which this is compensated. Moreover, development of the zygote with which high energy effectiveness and high power density are obtained also under a fuel and the high service condition of an air utilization rate as a technical problem to utilization is demanded.

[0005] Especially in the service condition of low operating temperature and the rate of high gas utilization, lock out (flooding) of the electrode porous body by condensation of a steam tends to take place in the air pole which water generates. In order to acquire a stable property over a long period of time, it is required, although the water repellence of an electrode is secured so that flooding may not happen. In especially the solid-state polyelectrolyte mold fuel cell from which high power density is obtained at low temperature, it is important.

[0006] In order to secure the water repellence of an electrode, it is effective that the content of the ion exchange group in ion exchange resin is small [of the ion exchange resin which covers a catalyst], namely, low in an electrode. However, since water content of ion exchange resin is low in this case, conductivity becomes low, and the cell engine performance falls. Furthermore, since the gas permeability of ion exchange resin falls, supply of the gas supplied to a catalyst front face through the covered ion exchange resin becomes slow. Therefore, the gas concentration in a reaction site falls, electrical-potential-difference loss becomes large, i.e., concentration overvoltage becomes high and an output declines.

[0007] For this reason, it is made to contain in an electrode, especially an air pole at the ion

exchange resin which covers a catalyst by making fluororesins, such as polytetrafluoroethylene (henceforth PTFE), tetrafluoroethylene / hexafluoropropylene copolymer, and tetrafluoroethylene / perfluoro (alkyl vinyl ether) copolymer, etc. into a water repellent agent, using resin with high ion exchange capacity, and the attempt which controls flooding is made (JP.5-36418.A). In addition, in this description, an A/B copolymer shows the copolymer which consists of a polymerization unit based on A, and a polymerization unit based on B.

[0008] However, if the amount of the above-mentioned water repellent agent in an electrode is made [many] in order to fully *****, as for the above-mentioned water repellent agent, the electric resistance of an electrode will increase for an insulator. Moreover, since the thickness of an electrode becomes thick, aggravation of gas permeability is caused, and there is a problem to which an output falls to reverse. In order to compensate conductive lowering of an electrode, it is required to raise the ion conductivity of the ion exchange resin which covers the conductivity of the carbon ingredient which is the support of a catalyst, and a catalyst. However, the electrode with which are simultaneously satisfied of sufficient conductivity and sufficient water repellence was difficult for obtaining, and was not easy to obtain high power and a polymer electrolyte fuel cell stable in the long run.

[0009] Moreover, although the approach (JP.7-211324.A) of mixing a fluoride pitch and the approach (JP.7-192738.A) of carrying out fluorination processing of the catalyst support are also proposed, there is a problem which cannot cover a catalyst front face with ion exchange resin to homogeneity. Moreover, the manufacture approach is complicated although the method (JP.5-251086.A, JP.7-134993.A) of giving inclination to water repellence to the thickness direction of an electrode is also proposed.

[0010]

[Problem(s) to be Solved by the Invention] In order to heighten an output, little direction of the amount of the water repellent agent contained in an electrode from a conductive and gas penetrable viewpoint is good. Moreover, the ion exchange resin which covers a catalyst is important when it raises the cell engine performance that conductivity is high and the permeability of gas is high, and its ion exchange resin with high exchange group concentration is desirable. However, when ion exchange resin with high exchange group concentration is used, the permeability and conductivity of fuel gas are high, and although the output in early stages of a fuel cell becomes high, if flooding tends to happen and it is used for a long period of time, lowering of an output will tend to take place.

[0011] Then, even if the gas permeability of this invention of the ion exchange resin which conductivity contains highly is high and uses it for a long period of time, it aims at offering the polymer electrolyte fuel cell which can maintain high power over a long period of time by having the air pole which has high water repellence.

[0012]

[Means for Solving the Problem] The gas diffusion electrode with which this invention contains a catalyst and ion exchange resin offers the solid-state polyelectrolyte mold fuel cell characterized by for said ion exchange resin contained in said air pole to be ion exchange resin which consists of a copolymer including the following polymerization unit A, the following polymerization unit B, and the following polymerization unit C in the solid-state polyelectrolyte mold fuel cell with which it is considered as the fuel electrode and the air pole, and said fuel electrode had been arranged at one side of a film-like solid-state polyelectrolyte, and said air pole has been arranged in another field, respectively:

Polymerization unit A: The polymerization unit based on the perfluoro vinyl ether which does not have the polymerization unit based on tetrafluoroethylene, the polymerization unit based on the perfluoro vinyl ether which has a polymerization unit B:sulfonic group, a polymerization unit C:ion exchange group, or the radical of the precursor.

[0013] In this invention, the ion exchange resin contained in an air pole is a copolymer which has a sulfonic group as an ion exchange group. It is desirable hydrolysis and to process acid type-ization and to obtain the precursor (only henceforth a precursor) which consists of resin which has -SO₂F which can change this copolymer into a sulfonic group. That is, it is desirable to carry out copolymerization of the perfluoro vinyl ether which has -SO₂F as a raw material as an

which conducts a proton] unit weight of an ion exchange group, and resistance of ion exchange resin becomes high.

[0024] moreover -- the ion exchange resin contained in an air pole -- the polymerization unit C -- the total amount of the polymerization unit A and the polymerization unit B -- receiving -- more than 2mol% -- being contained is desirable. The polymerization unit C of the role which raises ion conductivity is small.

[0025] However, it is thought that the gas which penetrates the ion exchange resin which covers a catalyst also penetrates the part of the ether linkage in ion exchange resin in addition to penetrating the water part of ion exchange resin. The polymerization unit C is included in ion exchange resin for the increase of an ether linkage part and the object which carries out and raises gas permeability in ion exchange resin. Therefore, if the content is too low, the reduction effectiveness of the concentration overvoltage by the improvement in gas permeability will not be accepted. As for the polymerization unit C, it is desirable that it is especially more than 5mol% to the total amount of the polymerization unit A and the polymerization unit B.

[0026] On the other hand, although especially the ion-exchange resin contained in the fuel electrode in this invention is not limited, fluorine-containing carbon sulfonic acid type ion-exchange resin is desirable. The ion exchange resin which consists of a copolymer which includes the polymerization unit A, the polymerization unit B, and the polymerization unit C like an air pole can also be used for a fuel electrode.

[0027] Since water does not generate with the reaction of a fuel cell in a fuel electrode, as long as it compares with an air pole, the content of the polymerization unit B and the polymerization unit C may be high. In order to secure the smooth fuel gas supply to a catalyst particle from a gaseous phase, the higher one of the above-mentioned content is good, and it is desirable that further especially the content of the polymerization unit B is high.

[0028] It is desirable that the catalyst included in the fuel electrode and air pole in this invention and ion exchange resin are catalyst-above-mentioned ion-exchange-resin =0.40:0.60-0.95:0.05 in a weight ratio from a viewpoint conductive [of an electrode], and water-repellent. In addition, in the case of the support catalyst with which the catalyst here was supported by support, such as carbon, the weight of this support shall be included.

[0029] It sets to this invention and an air pole and a fuel electrode (collectively henceforth an electrode) can form the liquid (henceforth the liquid for electrode formation) which dissolved or distributed ion exchange resin and a catalyst to the solvent by well-known approaches, such as fuel spray, spreading, and filtration. An electrode may be directly formed on an ion exchange membrane, and after forming in the shape of a layer on the charge collector which consists of carbon paper etc., it may join this to an ion exchange membrane. The sheet in which the carbon layer which consists of carbon and a fluororesin instead of carbon paper was formed on carbon fiber textile fabrics may be used. Moreover, an electrode may be formed on the plate prepared separately and this may be imprinted to ion exchange membrane. When not forming an electrode directly on ion exchange membrane, it joins to ion exchange membrane by well-known hot pressing, the pasting-up method (JP.7-220741.A, JP.7-254420.A), etc.

[0030] The desirable range changes with formation approaches of an electrode, and the thing of the viscosity range large from the thing of the shape of dispersion liquid which is dozens cp extent to the thing of the shape of a paste of 20000cp extent can be used for the viscosity of the liquid for the above-mentioned electrode formation. In order to adjust viscosity, the thickener and the diluent solvent may be contained in the liquid for electrode formation. Although the thing of ethyl cellulose, methyl cellulose, or a cellosolve system can be used as a thickener, not to use, since clearance actuation is needed is more desirable. As a diluent solvent, alcohols, such as a methanol, ethanol, and isopropyl alcohol, fluorocarbon, hydronalium fluorocarbon, hydronalium chlorofluorocarbon, water, etc. can be used.

[0031] In this invention, if a water repellent agent is contained in an electrode, especially an air pole, since the depressor effect over flooding will increase further, it is desirable. As a water repellent agent, tetrafluoroethylene / hexafluoroethylene copolymer, tetrafluoroethylene / perfluoro (alkyl vinyl ether) copolymer, PTFE, etc. can be used, for example. Moreover, since the fluorine-containing resin which can dissolve in a solvent tends to carry out water-repellent

approach of making said copolymer containing the polymerization unit B, to compound a precursor, and to acid-type--ization[hydrolysis and]-process this precursor.

[0014] As perfluoro vinyl ether which has these -SO2F, the perfluoro vinyl ether compound (m is the integer of 1-12 and p of the integer of 0-3 and n] 0 or 1 among a formula, and X is F or CF3) expressed with CF2=CF-(OCF2CFX) m-Op-(CF2) n-SO2F is desirable. The following compounds are mentioned as a desirable example of the above-mentioned perfluoro vinyl ether compound. However, among the following type, in q, the integer of 1-8 and r show the integer of 1-8, and s shows the integer of 1-3.

[0015] [Formula 1] CF2=CFO(CF2) qSO2F, CF2=CFOCF2CF(CF3) O(CF2) rSO2F, CF2=CF(OCF2CF(CF3)) sO(CF2)2SO2F.

[0016] Moreover, in this invention, the polymerization unit C included in the ion exchange resin contained in an air pole does not have the radical (radical of the precursor of an ion exchange group) which can turn into an ion exchange group by ion exchange groups, such as a sulfonic group and a phosphonic acid radical, or hydrolysis. As a polymerization unit C, the polymerization unit based on the perfluoro vinyl ether compound expressed with CF2=CF-(OCF2CFY) t-O-Rf is desirable. However, t is the integer of 0-3 among a formula, and Y is F or CF3. Rf is a perfluoroalkyl radical (1<u<=12) expressed with CuF2u+1 of a straight chain or branched chain.

[0017] The following compounds are mentioned as a desirable example of a perfluoro vinyl ether compound expressed with CF2=CF-(OCF2CFY) t-O-Rf. However, among the following type, in v, the integer of 1-8 and w show the integer of 1-8, and x shows the integer of 1-3.

[0018] [Formula 2] CF2=CFO(CF2) vCF3 and CF2=CFOCF2CF(CF3) O(CF2) wCF3, CF2=CF(OCF2CF(CF3)) xO(CF2)2CF3.

[0019] In this invention, although the ion exchange resin contained in an air pole consists of a copolymer including the polymerization unit A, the polymerization unit B, and the polymerization unit C, as long as the polymerization unit based on non-fluorine system olefins, such as a polymerization unit based on hexafluoropropylene and a fluoro olefin like chlorotrifluoroethylene, and ethylene, a vinylidene chloride, is less than [10mol%], in addition to the above-mentioned polymerization units A, B, and C, it may be included in this copolymer to all the polymerization units that constitute an air pole.

[0020] After -SO2F are hydrolyzed with water solutions, such as NaOH and KOH, are-izing [a precursor / an acid type] with water solutions, such as a hydrochloric acid and a sulfuric acid, and it is changed into acid type resin. For example, when hydrolyzing with a KOH water solution, -SO2F are changed into -SO3K, and target ion exchange resin is obtained in K ion being permuted by the proton after that. Moreover, hydrolysis and acid type-ized processing may be performed simultaneously.

[0021] As for the polymerization unit B and the polymerization unit C, in the ion exchange resin contained in an air pole, it is desirable that the total amount (mol) is contained to the polymerization unit A so that it may become [10-25-mol%]. Since, as for ion exchange resin, water content becomes low that it is less than [10mol%], conductivity becomes low and gas permeability also becomes low. Therefore, an electrode is formed into high resistance or an overvoltage becomes high. Moreover, if 25-mol% is exceeded, water content will become high too much, ion exchange resin will be swollen, and wettability will increase. Therefore, bywater, the pore of an air pole is easy to be blockaded and flooding tends to happen.

[0022] Since water generates with the reaction of a fuel cell in an air pole, especially buildup of the water content of ion exchange resin poses a problem. Moreover, since the gas continuously supplied to an air pole is usually humidified in order to prevent too much desiccation, it must also take into consideration buildup of the water content of the ion exchange resin by humid gas. the total amount of the polymerization unit B and the polymerization unit C over the polymerization unit A -- especially -- 12-22-mol% and further 15-20-mol% -- being contained is desirable.

[0023] Furthermore, on the ion exchange resin contained in an air pole, it is desirable that the polymerization unit B is more than 5mol% to the total amount of the polymerization unit A and the polymerization unit C. Less than [5mol%], there are few contents per [which has the role

treatment of the electrode, it is desirable.

[0032] Although it is usually desirable to be contained 0.01 to 30% of the weight in an electrode as for a water repellent agent, even if the amount of the water repellent agent which is an insulating material is little compared with the former, water repellence equivalent to the former is acquired by the air pole in this invention. Therefore, while being able to control resistance lifting of the electrode by addition of a water repellent agent to the minimum, there are also few possibilities that the pore of an electrode may be crushed by the water repellent agent.

[0033] Although especially the film-like solid-state polyelectrolyte in this invention is not limited, it is desirable to consist of resin which has a sulfonic group, a phosphonic acid radical, or a phenol system hydroxyl group as a cation-exchange radical for example. It is desirable to consist of fluorine-containing carbon sulfonic acid type ion exchange resin especially. The fluorine-containing carbon sulfonic acid type ion exchange resin which consists of a copolymer which specifically includes the polymerization unit A and the polymerization unit B may be illustrated.

and the polymerization unit C may be further included in said copolymer.

[0034] A film-like solid-state polyelectrolyte is fabricated by well-known approaches, such as heat press forming, roll forming, and extrusion molding, in the shape of film, and it hydrolyzes, it acid-type-ization-processes the precursor of said resin which has heat flow rate kinesis, and a film-like solid-state polyelectrolyte is obtained. Moreover, fluorine system cation exchange resin can also be obtained from the solution which dissolved in solvents, such as alcohol, by the solvent cast method.

[0035] Furthermore, a film-like solid-state polyelectrolyte may consist of ion exchange resin, such as hydrocarbon system resin which has a sulfonic group or a phosphonic acid radical, or hydrocarbon system resin by which partial fluorination was carried out. It may consist of resin which sulfonated the resin which introduced the sulfonic group into the polymerization unit based on styrene, polysulfone, a polyether ether ketone, etc., after specifically carrying out the graft polymerization of the styrene to ethylene / tetrafluoroethylene copolymer.

[0036] Moreover, a film-like solid-state polyelectrolyte may consist of film which compound-sized the above-mentioned cation exchange resin with reinforcing materials. As reinforcing materials, polyethylene, polytetrafluoroethylene, tetrafluoroethylene / perfluoro (propyl vinyl ether) copolymer, tetrafluoroethylene / hexafluoropropylene copolymer, etc. are mentioned. These reinforcing materials are used with the shape of the shape of fibril, and textile fabrics, the shape of a nonwoven fabric, and the gestalt of a porous body.

[0037] As for the thickness of a film-like solid-state polyelectrolyte, a 10-300-micrometer thing is used. When thinner than 10 micrometers, there is a possibility that it may be easy to generate a pinhole and it may short-circuit. If thicker than 300 micrometers, membranous electric resistance will become high and the output characteristics of a fuel cell will decline. The thickness of 20-100 micrometers is especially desirable.

[0038] Since the polymerization unit C is included in addition to the polymerization unit B, even if water generates the ion exchange resin contained in an air pole with the reaction of a fuel cell, resin is not swollen too much and wettability does not become high, either. Therefore, since the pass which gas diffuses can be secured since generation water is easy to be discharged, and many ether linkage is included, gas permeability of ion exchange resin is high. Therefore, even if it uses a fuel cell by the high current consistency, flooding of an electrode cannot happen easily, and concentration overvoltage can be made small. That is, electrical-potential-difference loss by diffusion of gas and supply being slow and the gas concentration in a reaction site falling can be made small.

[0039] [Example] Although an example (Examples 1-5) and the example of a comparison (Examples 6-7) explain this invention in detail below, this invention is not limited to these.

[0040] To the autoclave made from stainless steel, as a polymerization solvent, CF₂ClCF₂CH₂Cl, the azobisisobutyronitril as a polymerization initiator, and CF₂=CF-OCF₂CF(CF₃)-OCF₂CF₂SO₂F (a perfluoro (3, 6-dioxo-4-methyl-7-OKUTENIRU) sulfonyl fluoride --) Hereafter, and it called it PSVE, CF₂=CF-OCF₂CF₂CF₃ (it is called PPVE perfluoro (propyl vinyl ether) and the following) was taught. Subsequently, after fully deaerating the inside of an

autoclave with liquid nitrogen, tetrafluoroethylene (henceforth TFE) was taught and the polymerization was started at 70 degrees C. The pressure in an autoclave was uniformly held by introducing TFE from the outside of a system during the polymerization. Unreacted TFE is purged 10 hours after, the polymerization was terminated, the obtained polymer solution was condensed and dried [washed and] with the methanol, and the TFE/PSVE/PPVE copolymer was obtained.

[0041] After hydrolyzing this copolymer and rinsing in the mixed water solution containing 30 % of the weight of dimethyl sulfoxide, and 15 % of the weight of KOH(s), perfluorocarbon-sulfonic-acid mold ion exchange resin was obtained by being immersed into a 1-N hydrochloric acid.

[0042] In addition, seven kinds of ion exchange resin with which the content of each polymerization unit serves as a value shown in Examples 1-7 of a table 1 was compounded by adjusting the pressure and PPVE concentration at the time of the amount of a polymerization initiator, and a polymerization. Moreover, the ion exchange resin used for a fuel electrode side compounded the TFE/PSVE copolymer (it is 83:17 at a mole ratio) by the same approach except having not taught PPVE in the polymerization method of the copolymer used for the air pole.

After hydrolyzing this by the same approach as the copolymer used for the air pole, it was used having processed acid type-ization.

[0043] Next, as it was set to 3:1 by the weight ratio, the ethanol system solvent was made to distribute the catalyst which supports platinum to carbon black powder and becomes it, and the ion exchange resin obtained as mentioned above so that platinum may be contained 40% of the weight, and it considered as the catalyst paste for an air pole and fuel electrode formation.

[0044] As a charge collector, the fuel electrode and the air pole carried out water-repellent treatment of the carbon paper (trade name: TGP-H -060, Toray Industries, Inc. make), and used it. Moreover, after kneading the mixture which consists of 60 % of the weight of carbon black powder, and 40 % of the weight of PTFE powder as a gaseous diffusion layer, the sheet which was rolled out and PTFE fibrillated at 100 micrometers in thickness and 70% of voidage was obtained and used.

[0045] The electrode sheet was formed because a fuel electrode and an air pole apply a catalyst paste to the above-mentioned gaseous diffusion layer and dry. At this time, the catalyst paste was applied so that the amount of the platinum contained in an electrode sheet might serve as 0.5 mg/cm². The electrode sheet started the fuel electrode and the air pole so that an effective electrode surface product might be set to 2.28cm.

[0046] Perfluorocarbon-sulfonic-acid mold ion exchange membrane (trade name: the deflection myon HR, the Asahi Glass Co., Ltd. make, ion-exchange-capacity 1.1 milliequivalent / gram desiccation resin, 50 micrometers of thickness) was used as a solid-state polyelectrolyte. The air pole and the fuel electrode made the field where the catalyst paste was applied turn and counter inside, by performing a hotpress, where ion exchange membrane is put between them.

joined the film to the electrode sheet and produced the zygote.

[0047] The above-mentioned zygote was inserted between two carbon paper which is a charge collector, and it installed in the measurement cell, and ordinary pressure (ata) and gas performed continuous running in the constant current of 0.5 A/cm² in hydrogen/air processing subsystem, and the cell temperature of 80 degrees C, measured the output voltage of a cell at any time, and measured the days taken for the output voltage of a cell to decline by 50mV rather than initial value. A result is shown in a table 1.

[0048]

[A table 1]

| | 含有率 (mol%) | | | 初期のセル電圧 (V) | | 60 mV 低下する日数 |
|----|------------|------|------|-------------|--|--------------|
| | TFE | PSVE | PFVE | 圧置 | | |
| 例1 | 81 | 14 | 6 | 0.07 | | 140日 |
| 例2 | 70 | 10 | 14 | 0.08 | | 182日 |
| 例3 | 84 | 9 | 7 | 0.04 | | 186日 |
| 例4 | 86 | 4 | 20 | 0.03 | | 198日 |
| 例5 | 00 | 8 | 26 | 0.05 | | 83日 |
| 例6 | 78 | 22 | 0 | 0.07 | | 13日 |
| 例7 | 89 | 11 | 0 | 0.00 | | 136日 |

[0049]
[Effect of the Invention] Since according to this invention it is hard to swell the ion exchange resin contained in an air pole with humid gas or generation water, and its water content is not high and gas permeability is high, the sag by flooding is controlled. Therefore, few solid-state polyelectrolyte mold fuel cells of degradation of output characteristics with the passage of time with high and output density are obtained.

[Translation done.]